The role of linear dispersion in plane-wave self-phase modulation*

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It is shown by computer calculation that linear dispersion and self-phase modulation can strongly couple to influence pulse propagation phenomena. Pulses become distorted temporally as they propagate, and, under certain circumstances, optical shocks can form on the leading edge (in time) of a pulse. Some interesting cases are considered for pulse propagation in CS_2. The pulse compression scheme of Fisher, Kelley, and Gustafson is modified to take self-dispersive effects into consideration.

Self-phase modulation refers to the spectral broadening that a plane-wave pulse undergoes while propagating through a material which exhibits an intensity-dependent index of refraction. Since the index of refraction is varying in time as the pulse passes, the pulse will accumulate some degree of frequency modulation, or "chirp". In traversing a sample of length l, the nonlinear phase change is proportional to $\int_0^l \delta n(t_R, z_R)\, dz_R$, where δn is the nonlinear index change and the retarded transformation is given by $t_R \equiv t - zn_0/c$, $z_R \equiv z$. Here n_0 is the linear index of refraction. In the absence of dispersion, δn is a function of t_R only, and the accumulated nonlinear phase is proportional to $\delta n(t_R)$. In this dispersionless approximation, [1] the pulse shape remains constant when propagation distances are less than the self-steepening distance. [2]

We present some results of a plane-wave computer calculation which includes the interplay between dispersion and self-phase modulation. [3] This interplay must be understood in order to accurately assess the values of the nonlinear coefficients from experiments. We find, for instance, that one cannot always rely upon the distance rate of spectral growth for such an estimate. Also, the need is increasing for generating arbitrary pulses shapes (in time) from those readily available. Our results show that these new considerations can be crucial for pulse compression schemes. Although calculated examples of self-phase modulation[1,4] have traditionally avoided the influence of material dispersion, there have been several attempts to incorporate these two effects. Knight and Peterson, [5] for example, find (for the Lorentz model of a dielectric) a formula for the crossing of characteristics. These authors do not predict resultant pulse shapes, nor do they attempt to find shock distances when the nonlinear response is transient. Zel'dovich and Sobel'man[6] have also addressed their attention to the closely related case of pulse shortening when the center frequency is on either side of a strong absorption line. Their formula for the shock distance is meant only to apply to the resonant steady-state case. They state that in most transparent media (n_2 and $d^2n_0/d\lambda^2$ both positive), there is no compression. Because of the possibility of a transient response, we find that a short spike can form. Concurrently with the writing of this letter, Shimizu has shown[7] that for pulses in CS_2 shorter than the Kerr orientational relaxation time, a shock can form on the leading edge. We find that shocks can also occur with longer input pulses.

In our calculation, the material of length l is broken into small segments. The pulse is carried through each segment twice; once as a nonlinear segment and once as a dispersive segment. [8] Thus we solve a sequence of trivial problems. To ensure convergence, segments are chosen sufficiently thin. During propagation through a thin *nonlinear* segment of thickness dz_R, the phase gathers the increment $\delta\phi$ determined by the following equation[1,4]:

$$\tau \frac{\partial \delta\phi}{\partial t_R} = -\left(\delta\phi - \frac{\omega_0 n_2 dz_R}{2c}\mathcal{E}^2\right), \tag{1}$$

where n_2 is the coefficient of the nonlinear index change, $\mathcal{E}$ is the electric field envelope, and τ is the orientational relaxation time (2 psec in the case of CS_2).

During propagation through a dispersive segment of length dz, the complex electric field expression is given by

$$\begin{aligned} E_{\text{out}}(t, z+dz) = {} & [\exp(-i\omega_0 t)/2\pi] \int\int_{-\infty}^{\infty} d\Delta\omega\, dt'\, \mathcal{E}_{\text{in}}(t', z) \\ & \times \{\exp i[\Delta\omega(t'-t) + \delta\phi(t', z) + dz(\omega/c)n(\omega)] \\ & \times \exp[-dz\,(\omega/c)\kappa(\omega)]\}. \end{aligned} \tag{2}$$

Here $n(\omega)$ and $\kappa(\omega)$ are the real and imaginary parts of the complex index of refraction, and $\Delta\omega \equiv \omega - \omega_0$. In an attenuating or amplifying material, both parts of the refraction index can be important, but in a transparent material, $\kappa(\omega) = 0$, and the index of refraction is a very smooth function of ω. In expanding $n(\omega)$, the first term which can reshape pulses is proportional to $\lambda^3(d^2n_0/d\lambda^2)(\Delta\omega)^2$. Here λ is the free-space wavelength (*not* the wavelength in the material). As a check for our calculation, δn was set to zero, and the pulse duration doubled at the distance predicted by a hand calculation. Although the stimulated Raman effect can also have an influence on plane-wave pulse propagation, it will not be considered here.

We report here the simulation of pulse propagation effects in CS_2. An empirical formula[9] for the index of refraction of CS_2 was doubly differentiated to find $\lambda^3(d^2n_0/d\lambda^2) = 0.0845$ μm at $\lambda = 1.06$ μm. Figure 1 shows our calculated evolution in CS_2 of a 5-psec (full $1/e$ intensity duration) 1.06-μm pulse with a peak intensity of 22 GW/cm^2. We chose n_2 to be 1.3×10^{-11} esu. We are not considering self-focusing[10] because the tendency will be minimized for a pulse with a smooth spatial profile and a diameter greater than ~ 1 cm. The asymmetric evolution we observe in Fig. 1 is due to the delayed nonlinear response ($\tau = 2$ psec). In the absence of dispersion, the shape would not have changed. The leading

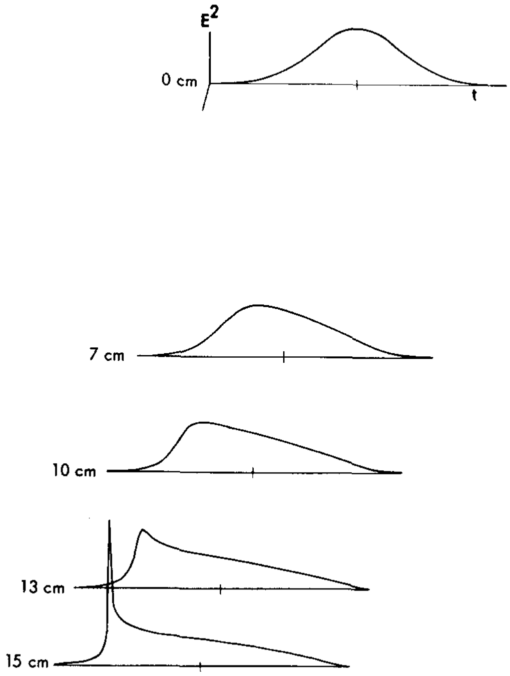

FIG. 1. Propagation of a 22-GW/cm^2 5-psec 1.06-μm pulse in CS_2. Note the formation of an optical shock on the leading edge.

edge steepening prior to shocking can be understood by considering (in the absence of dispersion) the plot of the instantaneous frequency shift versus time. There are two times where the "chirp" is the right sign for compression by normal (nonanomalous) dispersion. Because of the delay in nonlinear response, the "dispersion-compressible" portion on the leading edge is approximately 2 psec closer to the peak of the pulse, and the corresponding portion of the trailing edge is approximately 2 psec farther from the intensity peak. This is why features begin to appear on the leading edge. There is also a simple explanation for the shock which suddenly forms on the leading edge after the pulse has traveled 15 cm. Once a feature becomes shorter than for Kerr relaxation time, the instantaneous frequency (in the absence of dispersion) becomes proportional to the negative of the pulse envelope shape. This condition generates a "dispersion-compressible" chirp far closer to the peak of the feature than it would have been located in the steady-state case. Thus CS_2 exhibits an instability to temporal amplitude variations. For the same initial condition as above, the deleterious effect of amplitude noise has been examined with initial peak-to-peak (intensity) ripples of 10%, 2%, and 0.04%. The 10% noise triples in 2 cm. The shock formation shown in Fig. 1 can only be recognized in the case of 0.04% ripple. In repeating the calculation of Shimizu[7] we find excellent pulse-shape agreement if we let our calculated pulse propagate another 15%. At present we are unable to explain this minor discrepancy.

We have also calculated the dispersive effects which would be encountered in chirping mode-locked cw dye laser pulses via self-phase modulation in CS_2-filled waveguides. Since the multimoding observed in preliminary experiments[11] is not easy to simulate, we focus our interest on the simpler problem of single-mode propagation in which material dispersion dominates the guide dispersion. If it becomes necessary to use single-mode fibers to avoid mode hopping, the influence of the guide dispersion could be incorporated as a reduced effective value for $\lambda^3(d^2n_0/d\lambda^2)$. Self-focusing[10] is assumed to be absent in these waveguide calculations. Since $\lambda^3(d^2n_0/d\lambda^2)$ at 0.59 μm is 2.67 times greater than it is at 1.06 μm, CS_2 dispersion in the visible will have more influence than it had in the infrared. We have calculated reshaping for 5-psec (full $1/e$ intensity duration) 0.59-μm pulses (considering only material dispersion) with peak powers of 100, 50, and 10 MW/cm^2. The attenuation was chosen as $\alpha^{-1} = 200$ cm. Results are shown in Fig. 2. The 100-MW/cm^2 pulse travels less than 1 m prior to shocking. As the 50-MW/cm^2 pulse shocks, attenuation prevents catastrophic reshaping and the pulse shape is quite *stable* in the region of the shock (180–260 cm). [In comparison, the case with no attenuation (Fig. 1) reshapes severely as it shocks.] The pulse spectrum has broadened approximately eight times by 240 cm, and then does not broaden any more. As the shock forms, the pulse "speeds up" because the transient conditions favor spectral broadening to the Stokes' side ($\Delta\omega < 0$). In the 10-MW/cm^2 case, the pulse

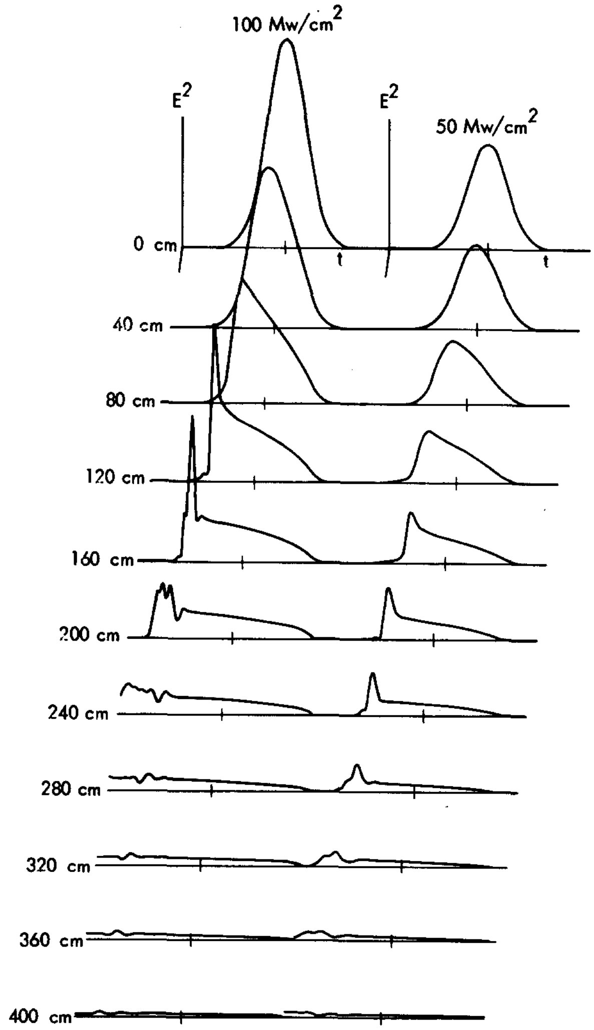

FIG. 2. Calculated temporal development of 0.59-μm 5-psec single-mode pulses in CS_2-filled waveguides. The attenuation was chosen so that low-level light would have its intensity reduced by $1/e$ every 2 m. Note the stabilizing role of the dissipation for the 50-MW/cm^2 case between 160 and 240 cm.

developed by 170-cm propagation to a skewed shape similar to the 50-MW/cm^2 case after 80 cm. The 10-MW/cm^2 case developed only twice the initial spectral broadening by 170 cm, and then retained its shape (except for size) and spectrum for the remaining 2.3-m propagation.

We have also improved the Fisher, Kelley, and Gustafson chirped-pulse grating-pair compression calculation[4] by including the effects of CS_2 dispersion. The pulse used in their example (1.06 μm, 22 GW/cm^2, 5 psec, 10 cm propagation) does change shape somewhat, and the chirp is considerably modified. In order to optimally compress this pulse, we required a grating pair with exactly *twice* the dispersive delay that was needed to compress the pulse in the dispersionless calculation. This is because the Stokes- (anti-Stokes) shifted light is generated early (late) in the pulse, and normal dispersion causes it to travel faster (slower). The chirp is *decreased* by this pulse-stretching effect. We have also calculated the grating-pair compressibility for the CS_2-filled waveguide cases. The 50-MW/cm^2 case has been studied after 2-m propagation. The portion which a grating pair would compress is several picoseconds past the shock feature, and we find that the compression is not nearly as critically dependent upon the dispersive delay setting of the grating pair as was the 22-GW/cm^2 10-cm CS_2 propagation case. Since the self-phase-modulated spectrum is far narrower in this subgigawatt case, the duration of the compressed pulse is reduced only to a $\frac{1}{2}$ psec. Although the "compressed" pulse does have approximately 6 times the peak power of the "uncompressed" pulse, the grating pair may not have been necessary because compression of the smooth portion of the pulse was at the expense of dispersing the already short shock.

In summary, this paper presents some results of a calculation which couples the propagation effects of both material dispersion and self-phase modulation. In CS_2, we find that shocks can form on the leading edge of pulses, and that pulses are quite unstable to amplitude noise. We also find corrections to the recipe for grating-pair compression of CS_2-chirped pulses. Knowledge of this correction factor is crucial for pulse compression because a 10% error in grating-pair setting is often unacceptable. Simulation studies of pulse propagation in CS_2-filled waveguides are also presented. It is found that if one wants to chirp pulses in CS_2-filled waveguides in order to later compress them via an appropriately adjusted grating pair, there is little advantage in choosing too long a sample. There are also cases where the shock may already be as short as that which the compressor could achieve. We would, for instance, recommend no more than 2-m propagation in CS_2-filled guides with 50-MW/cm^2 5-psec 0.59-μm pulses. Detailed examples of all the above cases will be the subject of a subsequent publication.

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[1] F. Shimizu, Phys. Rev. Lett. 19, 1097 (1967). See also, T.K. Gustafson, J-P. E. Taran, H.A. Haus, J.R. Lifsitz, and P.L. Kelley, Phys. Rev. 177, 306 (1969).
[2] F. DeMartini, C.H. Townes, T.K. Gustafson, and P.L. Kelley, Phys. Rev. 164, 312 (1967).
[3] R.A. Fisher and W. Bischel, Bull. Am. Phys. Soc. 18, 657 (1973).
[4] R.A. Fisher, P.L. Kelley, T.K. Gustafson, Appl. Phys. Lett. 14, 140 (1969).
[5] B.W. Knight and G.A. Peterson, Proc. Natl. Acad. Sci. U.S. 65, 841 (1970).
[6] B. Ya. Zel'dovich and I.I. Sobel'man, Zh. Eksp. Teor. Fiz. Pis'ma Red. 13, 182 (1971) [JETP Lett. 13, 129 (1971)].
[7] F. Shimizu, IEEE J. Quantum Electron. QE-8, 851 (1972).
[8] F.D. Tappert and C.N. Judice [Phys. Rev. Lett, 29, 1308 (1972)] and R.H. Hardin and F.D. Tappert [SIAM Rev. (Chronicle) 15, 423 (1973)] refer to the use of the "split-step algorithm" for the numerical solution of an extended Korteweg—de Vries equation. We suspect that their algorithm may be similar to the technique which we present in this letter.
[9] *International Critical Tables of Numerical Data, Chemistry, and Technology*, edited by E.W. Washburn (McGraw-Hill, New York, 1930).
[10] P.L. Kelley, Phys. Rev. Lett. 15, 1005 (1965).
[11] C.V. Shank, E.P. Ippen, and T.K. Gustafson, IEEE/OSA Conference on Laser Engineering and Application, Washington, D.C., 1973, Paper No. 12.6 (unpublished).